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CITATION:

Makita, Tadashi ...[et al]. Evaluation and correlation of viscosity data : the most probable values of the viscosity of gaseous ethane and ethylene. The Review of Physical Chemistry of Japan 1975, 44(2): 98-111

ISSUE DATE:

1975-04-30

URL:

<http://hdl.handle.net/2433/47004>

RIGHT:

THE REVIEW OF PHYSICAL CHEMISTRY OF JAPAN, VOL. 44, NO. 2, 1974

## EVALUATION AND CORRELATION OF VISCOSITY DATA

### The Most Probable Values of the Viscosity of Gaseous Ethane and Ethylene

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AND AKIRA NAGASHIMA\*\*

The critical evaluations of the viscosity data of gaseous ethane and ethylene have been carried out based on the experimental data available in the literatures. All the experimental measurements under high pressure were critically evaluated in view of their reliability, and the data were correlated with temperature and pressure. The most probable values of the viscosity are presented in the form of numerical tables covering the range of temperature from 298.15 to 473.15 K and that of pressure up to  $700 \times 10^5$  Pa for ethane, and from 297.15 to 373.15 K and up to  $800 \times 10^5$  Pa for ethylene. The estimated uncertainties of the tabulated values are also given in the table. The relation between residual viscosity and density has been also examined over the whole range of temperature and pressure.

#### Introduction

In the previous paper<sup>1)</sup>, we reported the results of the evaluation and correlation of the viscosity data of methane under high pressures. The present work is one of the successive programs of "High Pressure Data Center of Japan" organized in the Society of Material Science, Japan, with the sponsorship of the Agency of Science and Technology. The following members attended several meetings for the discussion concerning this work:

J. Osugi, Y. Takezaki (Kyoto Univ.); H. Iwasaki, S. Takahashi, K. Date (Tohoku Univ.);

I. Tanishita (Nippon Univ.); K. Watanabe (Keio Univ.)

to whom the authors wish to express sincere gratitude for their valuable suggestions and discussions.

#### Survey and Evaluation of Viscosity Data

There exist five and seven different measurements on the viscosity of gaseous ethane and ethylene under high pressures, respectively. In Table 1, the first author's names, methods of measurements, temperature ranges, and maximum pressures are listed in the order of the publishing year.

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(Received September 20, 1974)

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1) T. Makita, Y. Tanaka and A. Nagashima, *This Journal*, **43**, 54 (1973)

## Evaluation and Correlation of Viscosity Data

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Table 1 Measurements of the viscosity of ethane and ethylene under high pressures

First Author	Year	Method	Temp. Range (K)	Max. Press. (10 <sup>5</sup> Pa)	Ref. No.
Ethane					
Smith	1943	Rolling Ball	288-473	345	2
Meshcheryakov	1954	Transpiration	258-523	810	3
Baron	1959	Transpiration	324-408	552	4
Eakin	1962	Transpiration	298-444	552	5
Carmichael	1963	Rotating cylinder	299-477	358	6
Ethylene					
Mason	1940	Oscillating disk	281-299	50	7
Comings	1941	Transpiration	313	135	8
Comings	1944	Transpiration	303-368	169	9
Felsing	1944	Oscillating disk	303-323	186	10
Gonikberg	1947	Oscillating disk	297	1013	11
Golubev	1953	Transpiration	297-423	811	12
Neduzhii	1968	Transpiration	193-297	41	13

The original papers were carefully read through and examined from the view point of the reliability of the reported data by the operations similar to the previous work for methane.

The final evaluation was performed by the Committee members and several researchers in this field as described above. As the results, the high pressure data of ethane reported by Eakin *et al.*<sup>5)</sup> were considered to be the most reliable and given the highest weight. The weight second to the above was given to both sets of data by Carmichael *et al.*<sup>6)</sup> and Meshcheryakov *et al.*<sup>3)</sup> No weight was given to the remainders<sup>2, 4)</sup> in the present analysis. For ethylene the two sets of data by Comings *et al.*<sup>9)</sup> and Golubev *et al.*<sup>12)</sup> were considered to be the most reliable and given the highest weight. The second to the above was given to the other three sets of data<sup>10, 11, 13)</sup>. No weight was given to the two earlier works<sup>7, 8)</sup>.

2) A. S. Smith and G. Brown, *Ind. Eng. Chem. Ind. Ed.*, **35**(6), 705 (1943)

3) N. V. Meshcheryakov and I. F. Golubev, *Trudy GIAP*, Nos. 3-4 (1954)

4) J. D. Baron, J. G. Roof and F. W. Wells, *J. Chem. Eng. Data*, **4**, 283 (1959)

5) B. E. Eakin, K. E. Starling, J. P. Dolan and R. T. Ellington, *J. Chem. Eng. Data*, **7**, 33 (1962)

6) L. T. Carmichael and B. H. Sage, *J. Chem. Eng. Data*, **8**, 94 (1963)

7) S. G. Mason and O. Maass, *Canad. J. Res.*, **18B**, 128 (1940)

8) E. W. Comings and R. S. Egly, *Ind. Eng. Chem.*, **33**, 1224 (1941)

9) E. W. Comings, B. J. Mayland and R. S. Egly, *Univ. of Illinois, Eng. Expt. Sta. Bull.*, No. 354 (1944)

10) W. A. Felsing and F. Blankenship, *Proc. Okla. Acad. Sci.*, **24**, 90 (1944)

11) M. G. Gonikberg and L. F. Vereshchagin, *Compt. Rend. Acad. Sci. URSS*, **55**, 801 (1947)

12) I. F. Golubev and V. A. Petrov, *Trudy GIAP*, No. 2, 5 (1953)

13) I. A. Neduzhii and Yu. I. Khmara, *Teplofiz. Kharakter. Veshchestv*, **1**, 153 (1968); *Israel Program for Scientific Translations, Jerusalem*, TT 69-55091, 153 (1970)

### Method and Results of Correlation

The original experimental data were extracted and the values of temperature, pressure and viscosity were reduced to the SI units as follows:

temperature,  $T$ , in K  
 pressure,  $P$ , in  $10^5$  Pa ( $=1$  bar  $=0.9869$  atm)  
 viscosity,  $\eta$ , in  $10^{-8}$  Pa·s ( $=10^{-7}$  poise)

The method of the correlation is the same as used in our previous correlation. The viscosity values at the common grid-points of temperature and pressure were obtained from the original data reported in every work. When the data reported were not at one of the common grid-points specified, the interpolation procedures were carried out along an isotherm or an isobar on a digital computer using the method of least squares. In this procedure the special precaution was paid in order to keep up the experimental accuracy for each original work. The mean value at each grid-point was calculated with the weights determined in the critical evaluation. The standard deviations were also calculated by the following equation:

$$\sigma_1 = \sqrt{\frac{\sum [\omega_i (\eta_i - \bar{\eta})^2]}{\sum \omega_i (n-1)}} \quad (1)$$

where,  $\omega_i$  = the weight given,

$\eta_i$  = the viscosity in the original work,

$\bar{\eta}$  = the weighted mean value,

$n$  = the number of data.

On the other hand, since thirteen and twenty-one sets of experimental data at the atmospheric pressure are available for the viscosity of gaseous ethane and ethylene respectively, another correlation was carried out using several sets of experimental viscosity data at the normal pressure independently of the high pressure data. The viscosity values were fitted to the following quartic equations as a function of temperature:

ethane:

$$\begin{aligned} \eta_0 = & 4.929126 \times 10^2 - 2.087547 T + 2.094073 \times 10^{-2} T^2 \\ & - 3.665728 \times 10^{-3} T^3 + 2.196923 \times 10^{-8} T^4 \end{aligned} \quad (2)$$

ethylene:

$$\begin{aligned} \eta_0 = & 4.246821 \times 10^2 - 2.187278 T + 2.715949 \times 10^{-2} T^2 \\ & - 5.494674 \times 10^{-3} T^3 + 3.773645 \times 10^{-8} T^4 \end{aligned} \quad (3)$$

where  $\eta_0$  is given in  $10^{-8}$  Pa·s ( $10^{-7}$  poise) and  $T$  the temperature in K. Eq. (2) is found to fit the experimental data for ethane between 220 K and 520 K with the standard deviation of 0.87% and the maximum of 1.9%. Eq. (3) also reproduces the data between 170 K and 470 K with the standard deviation of 1.0% and the maximum of 2.6% for ethylene. The departures of the original data and several

correlated values from the above equations are plotted in Figs. 1 and 2. The weighted mean values at  $1.01 \times 10^5$  Pa have been compared with the correlated values obtained and are found to fit them with the mean deviation of 0.54% and the maximum of 1.18% for ethane, and with the mean deviation of 0.77% and the maximum of 1.40% for ethylene. Therefore the weighted mean values at low pressures

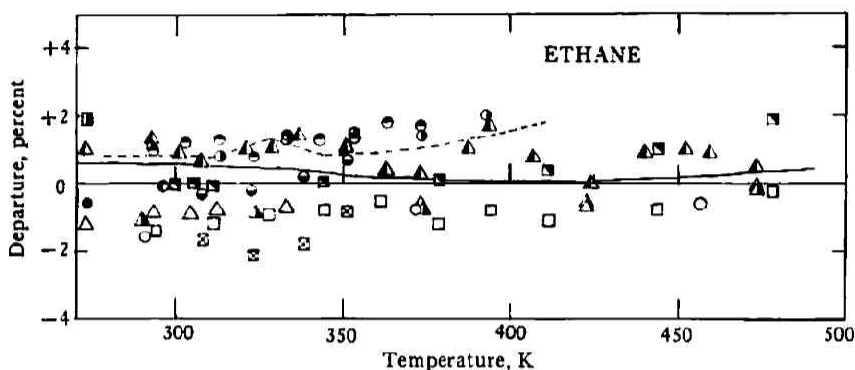


Fig. 1 Departure plots of viscosity values of ethane at the atmospheric pressure in literatures

$\triangle$ : (3),	$\bullet$ : (16),	$\boxtimes$ : (21),
$\square$ : (5),	$\circ$ : (17),	$\ominus$ : (22),
$\blacksquare$ : (6),	$\blacktriangle$ : (18),	$\blacktriangle$ : (23),
$\circ$ : (14),	$\bullet$ : (19),	--- : (24),
$\bullet$ : (15),	$\blacksquare$ : (20),	— : (25),

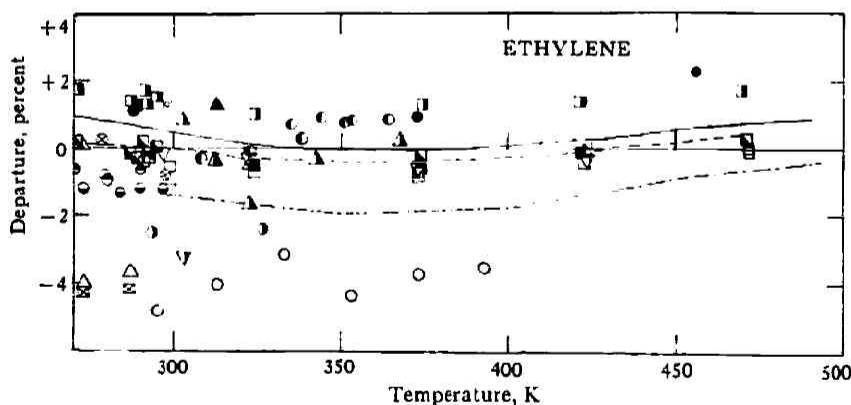


Fig. 2 Departure plots of viscosity values of ethylene at the atmospheric pressure in literatures

$\blacktriangle$ : (8),	$\bullet$ : (22),	$\blacksquare$ : (31),	$\boxtimes$ : (38),
$\blacktriangle$ : (9),	$\bullet$ : (26),	$\circ$ : (32),	$\nabla$ : (39),
$\nabla$ : (10),	$\bullet$ : (27),	$\blacktriangle$ : (33),	— : (25),
$\nabla$ : (12),	$\triangle$ : (28),	$\square$ : (34),	--- : (40),
$\bullet$ : (13),	$\boxtimes$ : (29),	$\ominus$ : (35),	--- : (41),
$\blacktriangle$ : (21),	$\blacksquare$ : (30),	$\odot$ : (37),	

Table 2 Weighted mean values of the viscosity of ethane in  $10^{-8}$  Pa·s ( $10^{-7}$  poise)

$P \times 10^5$ Pa	$T$ (K) (°C)											
	298.15	323.15	348.15	373.15	398.15	423.15	448.15	473.15				
	25	50	75	100	125	150	175	200				
1	934	1008	1080	1151	1220	1286	1350	1411				
5	946	1016	1087	1158	1227	1293	1359	1423				
10	962	1028	1097	1168	1237	1303	1372	1437				
20	999(11)	1055(4)	1120(10)	1192(10)	1260(9)	1326(12)	1397(12)	1463(17)				
30	1049(36)	1104(9)	1156	1226(12)	1293(12)	1357(17)	1423(18)	1486(23)				
40	1201(133)	1182(8)	1215(13)	1269(13)	1331(14)	1393(19)	1450(23)	1511(29)				
50	3973(9)*	1319(18)	1298(12)	1325(11)	1377(14)	1433(20)	1483(26)	1539(32)				
60	4393(52)	1603(28)	1418(5)	1396(7)	1433(14)	1480(20)	1520(26)	1571(35)				
70	4690(99)	2161(38)	1585(19)	1487(6)	1502(14)	1534(19)	1565(24)	1608(36)				
80	4927(104)	2896(89)	1809(46)	1604(12)	1579(13)	1595(20)	1617(23)	1651(37)				
90	5117(118)	3418(132)	2093(58)	1740(17)	1671(12)	1665(20)	1677(22)	1698(38)				
100	5337(167)	3828(97)	2512(16)	1934(11)	1763(11)	1737(19)	1744(22)	1748(38)				
120	5640(163)	4252(81)	3069(16)	2356(11)	2040(10)	1929(15)	1890(30)	1862(48)				
140	5907(189)	4609(87)	3504(48)	2759(4)	2336(9)	2138(9)	2048(33)	1981(62)				
160	6178(191)	4927(92)	3873(67)	3104(19)	2620(5)	2357(5)	2215(28)	2118(58)				
180	6424(209)	5207(104)	4189(74)	3416(30)	2892(6)	2577(4)	2386(25)	2260(53)				
200	6660(224)	5462(111)	4467(76)	3700(36)	3152(14)	2793(8)	2558(24)	2404(52)				
250	7212(253)	6023(119)	5033(74)	4307(45)	3732(27)	3306(22)	2992(32)	2775(54)				
300	7714(269)	6518(123)	5558(77)	4808(50)	4221(30)	3765(27)	3415(45)	3149(64)				
350	8176(276)	6971(127)	6001(81)	5244(56)	4654(38)	4179(34)	3799(50)	3505(73)				
400	8608(275)	7368(228)	6402(166)	5623(110)	5031(45)	4540(62)	4128(44)	3791(110)				
450	9014(225)	7779(246)	6801(197)	6000(136)	5382(50)	4879(65)	4453(45)	4096(108)				
500	9403(262)	8151(248)	7143(199)	6354(161)	5710(52)	5194(63)	4758(42)	4388(106)				
550	9781(255)	8550(276)	7531(236)	6679(170)	6029(57)	5499(66)	5057(45)	4685(116)				
600	10150(251)	8941(306)	7914(271)	7037(206)	6341(63)	5781(62)	5323(41)	4945(107)				
650	10517(254)	9314(329)	8275(292)	7371(225)	6650(68)	6066(68)	5593(42)	5208(109)				
700	10883(259)	9648(332)	8583(281)	7679(226)	6964(79)	6365(91)	5877(55)	5481(129)				

\* The values enclosed by dotted lines are of liquid phase.

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Table 3 Weighted mean values of the viscosity of ethylene in  $10^{-8}$  Pa·s ( $10^{-7}$  poise)

$P$ $10^5$ Pa	$T$ (K) (°C)		297.15		303.15		313.15		323.15		348.15		373.15	
			24		30		40		50		75		100	
1			1025		1046		1079		1111		1191		1267	
5			1032		1051		1087		1120		1199		1270	
10			1041		1059		1098		1132		1210		1275	
20			1068(4)		1081(13)		1125(3)		1160(4)		1232(9)		1289(12)	
30			1126(0)		1127(11)		1160(1)		1193(3)		1257(2)		1316(10)	
40			1209(9)		1191(10)		1212(2)		1238(8)		1279(18)		1348(4)	
50			1393*		1316(20)		1304(5)		1304(7)		1325(24)		1394(4)	
60			1788		1544(54)		1459(22)		1401(2)		1409(8)		1446(5)	
70			2742		1974(125)		1697(43)		1538(5)		1487(8)		1505(6)	
80			3304		2524(27)		1970(13)		1725(10)		1586(6)		1573(9)	
90			3637		3019(24)		2384(75)		1968(6)		1693(12)		1668(31)	
100			3896(24)		3412(13)		2801(38)		2333(21)		1830(18)		1751(29)	
120			4264(35)		3922(44)		3309(0)		2873(31)		2188(54)		1972(27)	
140			4605(37)		4303(30)		3778(0)		3311(47)		2561(80)		2251(4)	
160			4922(32)		4596		4123		3746(2)		2962		2509	
180			5216(20)		4877		4401		4060(14)		3217		2759	
200			5490(4)		5159		4699		4253(107)		3574		2996	
250			6099(48)		5691		5252		4911		4141		3540	
300			6622(113)		6178		5716		5349		4577		4022	
350			7083(174)		6590		6123		5745		4972		4452	
400			7502(227)		7078		6608		6137		5363		4838	
450			7894(264)		7407		6971		6522		5698		5189	
500			8272(288)		7762		7309		6854		6080		5512	
550			8642(298)		8178		7750				6442		5814	
600			9007(302)		8529		8158				6800		6102	
650			9366(306)		8801		8411				7053		6379	
700			9723(336)		9206		8808				7379		6652	
750			10048(382)		9499		9112				7661		6925	
800			10331(467)		9790		9385				7945		7200	

\* The values enclosed by the dashed lines are single point values which were determined by only one experimental data available in the literature.

Table 4 Coefficients of Equation (4)

Coefficient	Substance	Ethane			Ethylene		
		Moderate pressure region	High pressure region		Moderate pressure region	High pressure region	
B <sub>00</sub>		1.232902 × 10 <sup>3</sup>	-6.342937 × 10 <sup>5</sup>		5.093999 × 10 <sup>4</sup>	-2.183974 × 10 <sup>4</sup>	
B <sub>01</sub>		1.966653 × 10 <sup>6</sup>	9.741516 × 10 <sup>8</sup>		-6.274331 × 10 <sup>7</sup>	3.128532 × 10 <sup>8</sup>	
B <sub>02</sub>		-1.728377 × 10 <sup>9</sup>	-5.482866 × 10 <sup>11</sup>		2.988066 × 10 <sup>10</sup>	-2.846187 × 10 <sup>11</sup>	
B <sub>03</sub>		5.046018 × 10 <sup>11</sup>	1.339938 × 10 <sup>14</sup>		-6.345443 × 10 <sup>12</sup>	8.808950 × 10 <sup>13</sup>	
B <sub>04</sub>		-5.141028 × 10 <sup>13</sup>	-1.198857 × 10 <sup>16</sup>		5.041387 × 10 <sup>14</sup>	-9.069816 × 10 <sup>15</sup>	
B <sub>10</sub>		9.225225 × 10 <sup>2</sup>	5.716386 × 10 <sup>3</sup>		8.766533 × 10 <sup>3</sup>	1.215007 × 10 <sup>3</sup>	
B <sub>11</sub>		-1.421255 × 10 <sup>6</sup>	-8.555725 × 10 <sup>6</sup>		-1.260590 × 10 <sup>7</sup>	-4.635472 × 10 <sup>6</sup>	
B <sub>12</sub>		8.261413 × 10 <sup>8</sup>	4.687603 × 10 <sup>9</sup>		6.749558 × 10 <sup>9</sup>	3.672672 × 10 <sup>9</sup>	
B <sub>13</sub>		-2.141283 × 10 <sup>11</sup>	-1.111610 × 10 <sup>12</sup>		-1.595718 × 10 <sup>12</sup>	-1.076614 × 10 <sup>12</sup>	
B <sub>14</sub>		2.085013 × 10 <sup>13</sup>	9.646906 × 10 <sup>13</sup>		1.406404 × 10 <sup>14</sup>	1.079865 × 10 <sup>14</sup>	
B <sub>20</sub>		-4.789352 × 10	-1.782134 × 10		4.363085 × 10	1.198576 × 10	
B <sub>21</sub>		7.545328 × 10 <sup>4</sup>	2.608409 × 10 <sup>4</sup>		-2.124637 × 10 <sup>2</sup>	-5.425001 × 10 <sup>3</sup>	
B <sub>22</sub>		-4.468394 × 10 <sup>7</sup>	-1.390989 × 10 <sup>7</sup>		-2.954086 × 10 <sup>7</sup>	-1.980256 × 10 <sup>6</sup>	
B <sub>23</sub>		1.178127 × 10 <sup>10</sup>	3.196995 × 10 <sup>9</sup>		1.325764 × 10 <sup>10</sup>	1.335808 × 10 <sup>9</sup>	
B <sub>24</sub>		-1.165238 × 10 <sup>12</sup>	-2.674026 × 10 <sup>11</sup>		-1.667744 × 10 <sup>12</sup>	-1.735837 × 10 <sup>11</sup>	
B <sub>30</sub>		8.024483 × 10 <sup>-1</sup>	2.284905 × 10 <sup>-1</sup>		-1.393387	-3.549861 × 10 <sup>-2</sup>	
B <sub>31</sub>		-1.283465 × 10 <sup>3</sup>	-3.263160 × 10		7.644359 × 10 <sup>2</sup>	3.210202 × 10	
B <sub>32</sub>		7.697243 × 10 <sup>5</sup>	1.686591 × 10 <sup>4</sup>		1.812792 × 10 <sup>5</sup>	-9.314709 × 10 <sup>3</sup>	
B <sub>33</sub>		-2.049692 × 10 <sup>8</sup>	-3.724269 × 10 <sup>6</sup>		-1.682783 × 10 <sup>8</sup>	7.899434 × 10 <sup>5</sup>	
B <sub>34</sub>		2.043200 × 10 <sup>10</sup>	2.956581 × 10 <sup>8</sup>		2.484619 × 10 <sup>10</sup>	2.267469 × 10 <sup>7</sup>	
B <sub>40</sub>		-2.545125 × 10 <sup>-3</sup>	-1.021520 × 10 <sup>-5</sup>		3.139115 × 10 <sup>-2</sup>	4.836930 × 10 <sup>-5</sup>	
B <sub>41</sub>		3.900140	1.417171 × 10 <sup>-2</sup>		-3.767291 × 10	-5.667450 × 10 <sup>-2</sup>	
B <sub>42</sub>		-2.217875 × 10 <sup>3</sup>	-7.037706		1.671119 × 10 <sup>4</sup>	2.484040 × 10	
B <sub>43</sub>		5.523379 × 10 <sup>5</sup>	1.468531 × 10 <sup>3</sup>		-3.241224 × 10 <sup>6</sup>	-4.845099 × 10 <sup>3</sup>	
B <sub>44</sub>		-5.051650 × 10 <sup>7</sup>	-1.072167 × 10 <sup>5</sup>		2.313886 × 10 <sup>8</sup>	3.560601 × 10 <sup>5</sup>	



(1, 5 and  $10 \times 10^5$  Pa) were slightly adjusted on the basis of the correlated values at the normal pressure by the graphical method so that the weighted mean values and the correlated values should be consistent with each other.

The final weighted mean values are tabulated in Table 2 for ethane and Table 3 for ethylene, respectively. The standard deviations are also given in the parentheses except for the adjusted values near the normal pressure and the single point values of ethylene. In this procedure every tabulated value under pressures was determined quite independently of the adjacent values. However the smoothness and the consistency among the values have been found rather fair for ethane, but not quite satisfactory for ethylene.

### Recommended Viscosity Values

The weighted mean values obtained above were smoothed using the following appropriate empirical equation, in which the viscosity is expressed as a function of both temperature and pressure:

$$\eta = \sum_{i=0}^4 B_{0i}/T^i + (\sum_{i=0}^4 B_{1i}/T^i)P + (\sum_{i=0}^4 B_{2i}/T^i)P^2 + (\sum_{i=0}^4 B_{3i}/T^i)P^3 + (\sum_{i=0}^4 B_{4i}/T^i)P^4 \quad (4)$$

Since the viscosity values of gaseous ethane and ethylene increase rather rapidly with pressure near the critical point\*, it was found to be impossible to fit the whole values to a single equation. Therefore the evaluated values for each gas were divided into two regions by appropriate pressures, namely the moderate pressure region ( $1 \sim \text{about } 110 \times 10^5$  Pa) and the high pressure region (about  $110 \sim 800 \times 10^5$  Pa). The boundary pressure varies slightly with temperature between  $100 \times 10^5$  Pa and  $120 \times 10^5$  Pa. The 25 empirical coefficients of Eq. (4) were determined for each region by the least squares method. The coefficients determined are given in Table 4. For Eq. (4)  $\eta$  is the viscosity given in  $10^{-8}$  Pa·s,  $T$  the temperature in K and  $P$  the pressure in  $10^5$  Pa (=bar). It has been found that the equation is able to reproduce the weighted mean values within the percentage standard deviations  $\sigma_2$  as follows:

$$\sigma_2 = 100 \sqrt{\frac{1}{n} \sum [(\bar{\eta} - \eta_{\text{calc}})/\eta_{\text{calc}}]^2}$$

where  $\bar{\eta}$  = weighted mean values at grid-points.

$\eta_{\text{calc}}$  = calculated values of the viscosity by Eq. (4).

$n$  = number of data points.

	Moderate pressure region ( $1 \sim 110 \times 10^5$ Pa)	High pressure region ( $110 \sim 800 \times 10^5$ Pa)
Ethane	0.31	0.42
Ethylene	0.58	0.65

The values of percentage standard deviation for each set of original data are summarized in Table 5. For some of the original data and the correlated values, the percentage departures from Eq. (4) were calculated by the following equation

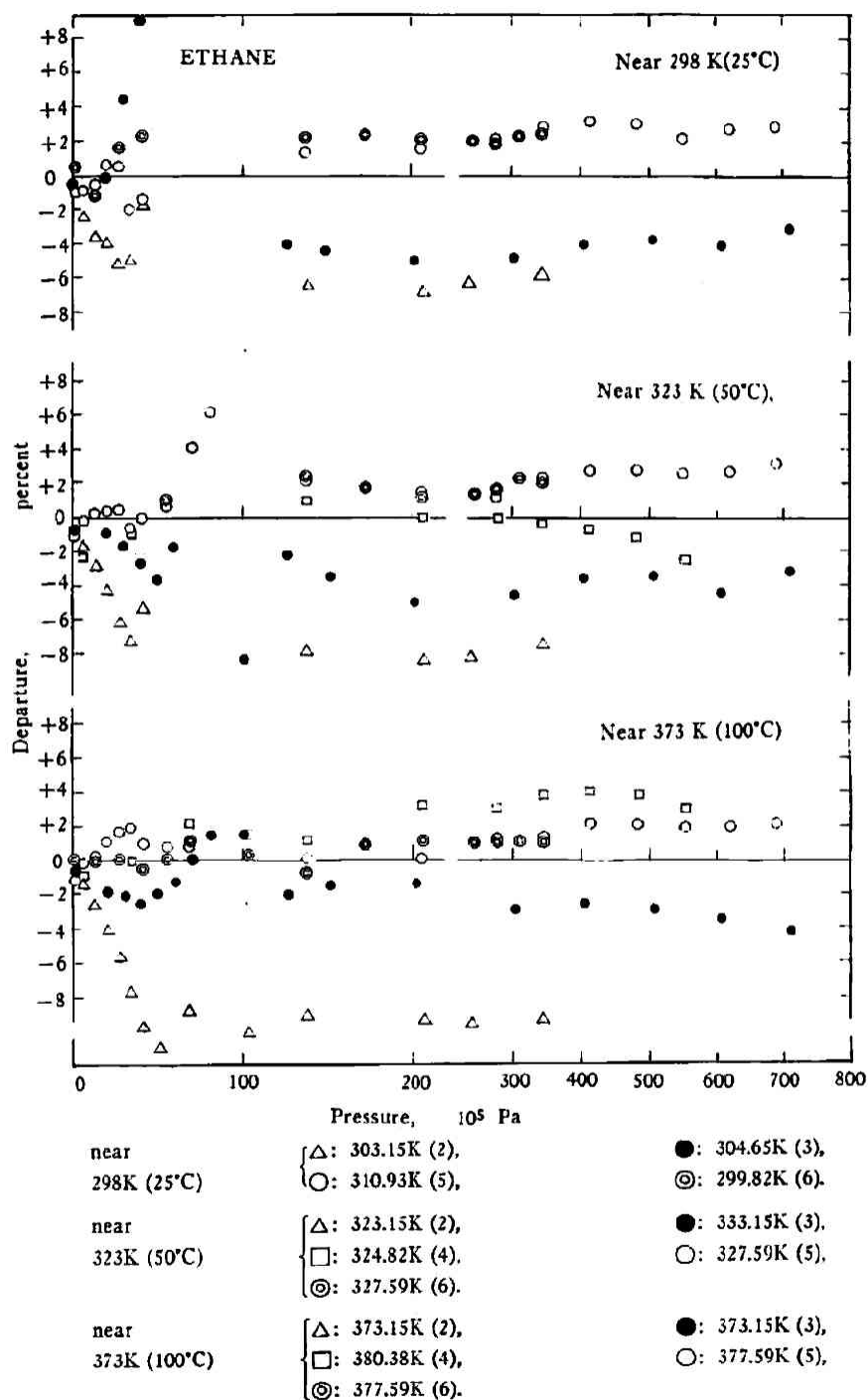


Fig. 3 Percentage departures of the original viscosity data for ethane under high pressures

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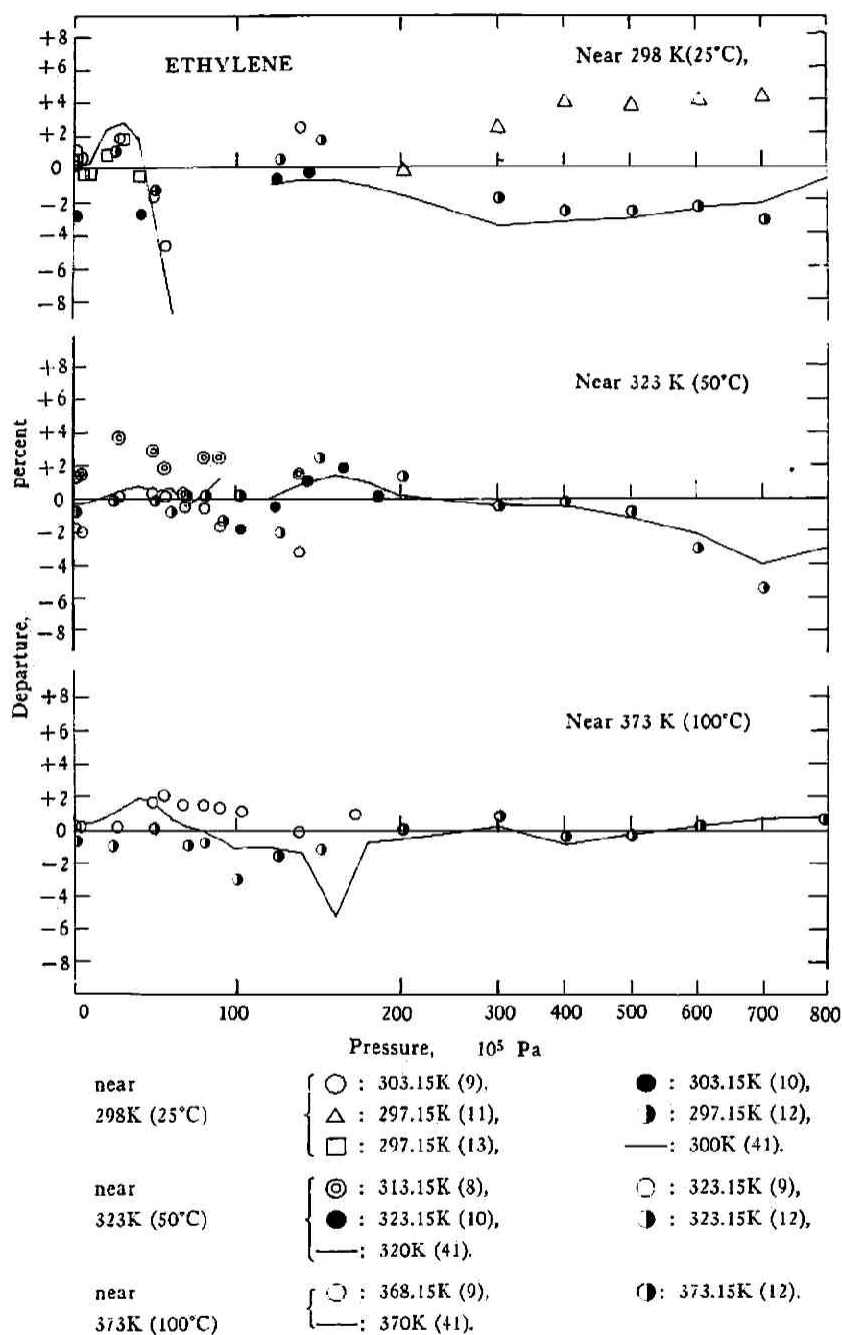


Fig. 4 Percentage departures of the original viscosity data for ethylene under high pressures

Table 5 Percentage standard deviation from Equation (4)

First Author	Moderate pressure region		High pressure region	
	Number of data points compared	Standard deviation percent	Number of data points compared	Standard deviation percent
<b>Ethane</b>				
Carmichael	41	1.08	56	1.74
Eakin	75	1.14	72	1.98
Baron	11	1.44	28	2.55
Meshcheryakov	44	3.82	54	4.35
Smith	81	5.96	40	10.7
<b>Ethylene</b>				
Neduzhii	6	0.87		
Golubev	17	1.04	27	2.36
Comings (1944)	36	1.40	5	2.67
Comings (1941)	8	2.35	1	1.50
Felsing	3	2.55	6	0.95
Gonikberg	1	36.2	7	3.72

$$\text{departure percent} = \frac{\eta_{lit} - \eta_{calc}}{\eta_{calc}} \times 100$$

where  $\eta_{lit}$  is the original viscosity data. The results are plotted as a function of pressure in Figs. 3 and 4. The final recommended viscosity values for gaseous ethane and ethylene have been generated by Eq. (4) and given in Tables 6 and 7, covering the temperatures from 300 to 475 K and pressures between 1 and  $700 \times 10^5$  Pa for ethane, and the temperatures from 300 to 380 K and pressures between 1 and  $800 \times 10^5$  Pa for ethylene, respectively. Eq. (4) is not adequate to reproduce the viscosity values precisely near the critical point as shown by the blanks of the recommended values in Tables 6 and 7.

### Residual Viscosity Correlation

As another trial, the residual viscosity  $\bar{\eta} - \eta_0$  was correlated with density by a simple polynomial equation as follows:

$$\bar{\eta} - \eta_0 = \sum_{i=1}^4 C_i \rho^i \quad (5)$$

where  $\bar{\eta}$  = weighted mean value of viscosity in  $10^{-8}$  Pa·s.

$\eta_0$  = viscosity in  $10^{-8}$  Pa·s calculated by equation (2) for ethane and (3) for ethylene.

$\rho$  = density in  $\text{g}\cdot\text{cm}^{-3}$ .

The values of density were calculated by modified BWR equations of state<sup>42)</sup>. The empirical coefficients  $C_i$  determined are listed in Table 8. As shown in Figs. 5 and 6 these correlations can be used in the range of density below  $2\rho_c$  ( $\rho_c$  is the critical density) except for narrow regions near the critical point with the estimated error of about 1.5% for ethane and 2% for ethylene.

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Table 6 Recommended values of the viscosity of gaseous ethane  
in  $10^{-8}$  Pa·s ( $10^{-7}$  poise)

Pressure $10^5$ Pa (bar)	Temperature, K							
	300	325	350	375	400	425	450	475
1	934	1014	1087	1157	1225	1291	1354	1415
10	974	1032	1101	1172	1241	1309	1376	1443
20	995	1057	1127	1196	1265	1333	1401	1468
30	1049	1102	1165	1230	1296	1362	1427	1491
40	1201	1185	1220	1274	1335	1396	1456	1515
50		1330	1300	1331	1381	1436	1490	1542
60		1565	1411	1401	1437	1482	1528	1573
70			1567	1488	1501	1535	1571	1610
80			1780	1598	1579	1598	1623	1652
90			2061	1732	1664	1665	1678	1699
100			2436	1902	1767	1742	1737	1753
120	5535*	4144	3020	2340	2009	1896	1873	1848
140	5814	4505	3418	2711	2318	2126	2042	1981
160	6071	4826	3774	3050	2603	2350	2207	2114
180	6322	5116	4095	3360	2872	2566	2379	2255
200	6557	5383	4384	3644	3126	2778	2550	2398
250	7101	5952	4999	4253	3687	3272	2973	2758
300	7583	6420	5490	4750	4168	3717	3372	3110
350	8041	6836	5906	5172	4587	4118	3747	3453
400	8460	7238	6290	5553	4961	4486	4092	3774
450	8853	7614	6659	5904	5315	4820	4418	4078
500	9249	8033	7016	6270	5631	5117	4704	4357
550	9632	8418	7403	6587	5925	5396	4974	4615
600	9973	8825	7804	6943	6267	5690	5249	4883
650	10341	9161	8123	7254	6530	5982	5504	5149
700	10697	9451	8418	7577	6890	6277	5779	5405

\* The values enclosed by dotted lines are of liquid phase.

\* The critical constants are as follows;

Ethane  $T_c = 305.43$  K,  $P_c = 48.80 \times 10^5$  Pa,  $V_c = 148$  cm<sup>3</sup>·mol<sup>-1</sup>Ethylene  $T_c = 282.36$  K,  $P_c = 50.32 \times 10^5$  Pa,  $V_c = 129$  cm<sup>3</sup>·mol<sup>-1</sup>14) E. Ziegler, *Phil. Diss. Halle Univ.*, 1 (1904)15) H. Vogel, *Ann. Physik*, **43**, 1235 (1914)16) Y. Ishida, *Phys. Rev.*, **21**, 550 (1923)17) T. Titani, *Bull. Chem. Soc. Japan*, **5**, 98 (1930)18) M. Trautz and K. G. Sorg, *Ann. Physik*, **10**, 81 (1931)19) H. Azumi, *Bull. Chem. Soc. Japan*, **12**, 199 (1937)20) V. D. Majumder and V. S. Oka, *J. Univ. Bombay*, **17 A**, Part 5, 35 (1949)21) P. M. Craven and J. D. Lambert, *Proc. Roy. Soc. London*, **A 205**, 439 (1951)22) J. D. Lambert, K. J. Cotton, M. W. Pailthorpe, A. M. Robinson, J. Scrivins, W. R. F. Vale and R. M. Young, *Proc. Roy. Soc. London*, **231 A**, 280 (1955)23) A. G. DeRocco and J. O. Halford, *J. Chem. Phys.*, **28**, 1152 (1958)

Table 7 Recommended values of the viscosity of gaseous ethylene  
in  $10^{-8}$  Pa·s ( $10^{-7}$  poise)

Pressure $10^5$ Pa (bar)	Temperature, K								
	300	310	320	330	340	350	360	370	380
1	1030	1067	1103	1136	1167	1196	1225	1257	1292
10	1055	1085	1122	1157	1187	1213	1238	1265	1297
20	1074	1107	1149	1184	1210	1231	1253	1280	1319
30	1109	1143	1184	1214	1238	1254	1272	1302	1351
40	1196	1203	1231	1257	1275	1287	1303	1333	1391
50	1370	1304	1304	1316	1326	1335	1346	1378	1436
60	1672	1473	1410	1399	1398	1398	1405	1429	1490
70		1731	1566	1510	1491	1483	1479	1497	1554
80		2104	1785	1660	1607	1579	1562	1570	1628
90			2092	1860	1757	1698	1662	1659	1726
100				2122	1939	1835	1764	1759	1853
120	4110	3548	3070	2679	2373	2153	2022	1979	2023
140	4443	3906	3456	3084	2777	2540	2369	2270	2242
160	4754	4232	3805	3445	3139	2886	2688	2543	2460
180	5044	4533	4118	3769	3463	3202	2979	2801	2680
200	5312	4804	4402	4056	3755	3484	3243	3046	2896
250	5908	5390	4997	4665	4359	4080	3823	3597	3422
300	6416	5879	5481	5148	4844	4556	4297	4081	3921
350	6863	6301	5894	5554	5242	4955	4702	4505	4381
400	7276	6691	6274	5926	5605	5313	5064	4883	4803
450	7655	7068	6636	6280	5948	5644	5389	5222	5177
500	8037	7435	7016	6645	6308	5978	5706	5536	5521
550	8408	7804	7408	7069	6690	6325	6006	5830	5837
600	8732	8163	7809	7458	7043	6651	6273	6100	6139
650	9099	8542	8215	7889	7458	6996	6600	6360	6436
700	9455	8906	8654	8291	7864	7331	6870	6615	6784
750	9776	9216	8979	8635	8171	7581	7095	6887	7177
800	10016	9456	9259	8957	8401	7771	7213	7104	7625

Table 8 Coefficients of equation (5)

	Ethane	Ethylene
$C_1$	$1.31666 \times 10^3$	$1.21667 \times 10^3$
$C_2$	$3.34167 \times 10^4$	$3.11667 \times 10^4$
$C_3$	$-9.66667 \times 10^4$	$-9.16667 \times 10^4$
$C_4$	$2.08333 \times 10^5$	$1.83333 \times 10^5$

The authors wish to thank Mr. M. Ikeda, graduate student at Keio Univ., Miss K. Hamaoka and Miss N. Konishi for their help in the data processing of this work.

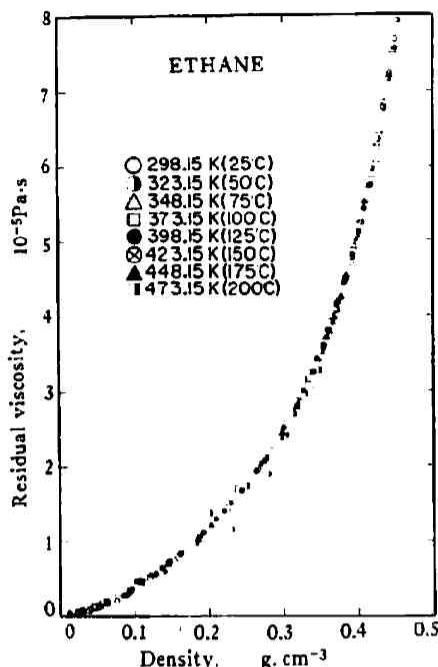


Fig. 5 Residual viscosity correlation for ethane

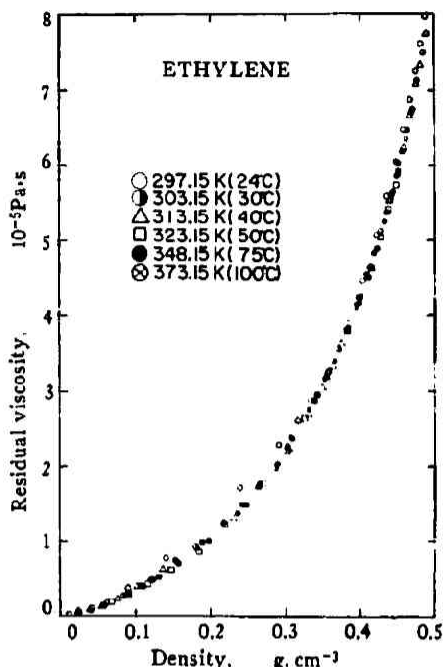


Fig. 6 Residual viscosity correlation for ethylene

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